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UB matrix implementation for inelastic neutron scattering experiments

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The UB matrix approach has been extended to handle inelastic neutron scattering experiments with differing k_i and k_f. We have considered the typical goniometer employed on triple-axis and time-of-flight spectrometers. Expressions are derived to allow for calculation of the UB matrix and for converting from observables to **Q**-energy space. In addition, we have developed appropriate modes for calculation of angles for a specified **Q**-energy position.

1. Introduction

Inelastic neutron scattering provides a very powerful tool for investigation of both magnetic and structural excitations in condensed matter systems. Although the field is rather mature, with the first instruments developed in the 1950s, handling of single crystals is in a rather primitive state. There are two standard instruments for investigations using this technique. First, the triple-axis spectrometer is the standard instrument used at rector-based neutron sources and consists of monochromator crystals before and after the sample, allowing one to study excitations with a specific wavevector and energy transfer. The standard method for handling crystals on such an instrument is to define a pair of orthogonal reciprocal-space vectors within a scattering plane and work within the space defined by these vectors. The second type of instrument, most often used at pulsed spallation neutron sources, uses measurement of the neutron time-of-flight with either a fixed incident or final wavevector to measure the neutron energy transfer. Such an instrument often employs very large detector banks to maximize the solid-angle coverage. Measurement with single crystals using these time-of-flight instruments is at a fairly early stage and very little software has been developed to assist in performing or analyzing such experiments. We have extended the UB matrix formalism (Busing & Levy, 1967) to handle such inelastic neutron scattering experiments and have performed calculations for the typical goniometers employed on such instruments. This paper will be split into two parts, the first dealing with the triple-axis spectrometer and the second with time-of-flight spectrometers.

2. Triple-axis spectrometer

The triple-axis spectrometer (TAS) was invented in the 1950s by Bertran Brockhouse (Brockhouse & Stewart, 1955) and remains the principal tool for studying inelastic neutron scattering at reactor-based continuous neutron sources. This instrument uses Bragg scattering from a monochromator and analyzer crystal to study scattering at a specified $\mathbf{k}_i$ and $\mathbf{k}_f$. Many of the early measurements were performed on high-symmetry single crystals and most of the data acquisition software worked under this assumption. Consequently, the typical way of defining an orientation for a single crystal on such an instrument would be to specify two orthogonal vectors within the scattering plane and to work in the coordinate space of these vectors. Such a procedure works well provided that the crystal symmetry is sufficiently high. However, measurements on lower-symmetry samples are becoming much more prevalent and a more flexible approach is clearly needed. In addition, despite the presence of a goniometer that allows some movement away from the scattering plane, the data acquisition schemes employed to date usually do not allow for such motions.

Clearly, the best approach to overcome these limitations is the standard UB matrix approach (Busing & Levy, 1967). To apply the UB matrix formalism to the TAS, the expressions need to be generalized to handle the inelastic scattering case where the incident and final wavevectors differ. In addition, we need to consider a different goniometer for the TAS as most of these instruments have sample manipulation which is provided by a series of three rotations, as shown in Fig. 1. These rotations consist of a single rotation about the instrument z axis, and a pair of arcs (lower and upper, angles μ and ν, respectively) with rather limited angular range (typically $\pm 25^\circ$). Following Busing & Levy (1967), the z-axis rotation is broken down into the angles ω and θ. The upper arc, ν, is defined to be a rotation about the x axis, while the lower arc, μ, is defined to be a rotation about the y axis. For all rotations, we use the standard trigonometric definition, positive rotations are counterclockwise, resulting in the following rotation matrices:

$$\mathbf{N} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\nu & -\sin\nu \\ 0 & \sin\nu & \cos\nu \end{pmatrix}, \qquad (1)$$

$$\mathbf{M} = \begin{pmatrix} \cos\mu & 0 & \sin\mu \\ 0 & 1 & 0 \\ -\sin\mu & 0 & \cos\mu \end{pmatrix}, \quad (2)$$

$$\mathbf{\Omega} = \begin{pmatrix} \cos\omega & -\sin\omega & 0 \\ \sin\omega & \cos\omega & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad (3)$$

$$\mathbf{\Theta} = \begin{pmatrix} \cos\theta & -\sin\theta & 0 \\ \sin\theta & \cos\theta & 0 \\ 0 & 0 & 1 \end{pmatrix}. \quad (4)$$

It is worth pointing out several differences between the inelastic and diffraction cases. First, the scattering angle has been designated by φ as opposed to 2θ. The reason for this designation, as will become clear below, is that the θ angle for the general inelastic case will not be half of the scattering angle. In addition, we will write the momentum transfer

$$\mathbf{Q} = \mathbf{k}_\mathrm{i} - \mathbf{k}_\mathrm{f} \quad (5)$$

as opposed to $\mathbf{Q} = \mathbf{k}_\mathrm{f} - \mathbf{k}_\mathrm{i}$ used by Busing & Levy (1967). This definition is adopted for consistency with the standard definition of energy transfer where neutron energy loss is positive, *i.e.* $\Delta E = E_\mathrm{i} - E_\mathrm{f}$.

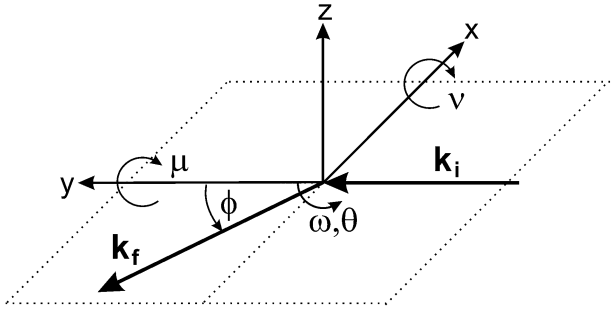

Figure 1
Diagram showing the sense of rotations of the angles of the triple-axis spectrometer with all others set to zero. The angle ν corresponds to the upper goniometer arc, μ to the lower arc, and the combination of ω and θ the sample rotation angle.

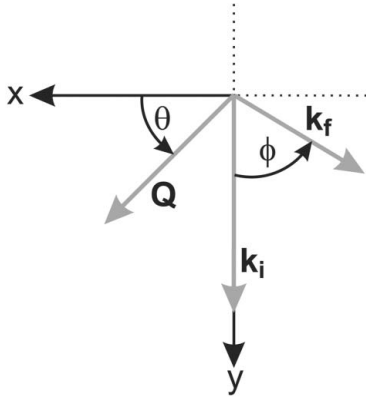

Figure 2
Diagram showing full inelastic momentum transfer in the laboratory coordinate system.

Like Busing & Levy (1967), we begin with a certain wavevector,

$$\mathbf{Q} = \begin{pmatrix} h \\ k \\ l \end{pmatrix}. \quad (6)$$

The **B** matrix transforms **Q** into the orthogonal crystal coordinate system ($\mathbf{Q}_\mathrm{c}$), which is then transformed by the **U** matrix into the coordinate system attached to the upper axis of the instrument where the crystal is mounted and thus remains stationary; in this case the ν coordinate system ($\mathbf{Q}_\nu$). The goal of the calculation is to observe this wavevector in the detector with a specified k_i and k_f. Using Fig. 2, we will begin by writing out the expression for **Q** in the laboratory coordinate system, $\mathbf{Q}_\mathrm{L}$, as

$$\mathbf{Q}_\mathrm{L} = \mathbf{\Theta\Omega MNUBQ}, \quad (7)$$

$$\begin{aligned} &= \mathbf{k}_\mathrm{i} - \mathbf{k}_\mathrm{f} \\ &= \begin{pmatrix} 0 \\ k_\mathrm{i} \\ 0 \end{pmatrix} - \begin{pmatrix} -k_\mathrm{f}\sin\varphi \\ k_\mathrm{f}\cos\varphi \\ 0 \end{pmatrix} \\ &= \begin{pmatrix} k_\mathrm{f}\sin\varphi \\ k_\mathrm{i} - k_\mathrm{f}\cos\varphi \\ 0 \end{pmatrix}. \end{aligned} \quad (8)$$

If we define q to be the length of the vector **Q**, which can be calculated in any orthogonal coordinate system, we can use the above expressions to write

$$q^2 = k_\mathrm{i}^2 + k_\mathrm{f}^2 - 2k_\mathrm{i}k_\mathrm{f}\cos\varphi. \quad (9)$$

Using equation (8) and Fig. 2, we can define the angle θ as

$$\tan\theta = (k_\mathrm{i} - k_\mathrm{f}\cos\varphi)/k_\mathrm{f}\sin\varphi. \quad (10)$$

Equations (9) and (10) represent the principal difference in the extension of the calculation to handle the fully inelastic case. The simple expressions for q and θ given by Busing & Levy (1967) need to be replaced by the equations above. With the above definition of θ, we can calculate **Q** in the θ coordinate system, $\mathbf{Q}_\theta$, by operating on $\mathbf{Q}_\mathrm{L}$ with $\mathbf{\Theta}^{-1}$, as

$$\mathbf{Q}_\theta = \mathbf{\Omega MNUBQ} = \begin{pmatrix} q \\ 0 \\ 0 \end{pmatrix}. \quad (11)$$

Note that this is identical to the expression defined by Busing & Levy (1967).

At this point, we can calculate a unit vector in the ν coordinate system. Note that this coordinate system plays the same role as the φ coordinate system of Busing & Levy (1967) in that it represents the uppermost axis of the instrument where the crystal remains fixed. If we set the wavevector length to unity, we calculate

$$\mathbf{u}_\nu = \mathbf{N}^{-1}\mathbf{M}^{-1}\mathbf{\Omega}^{-1}\mathbf{u}_\theta = \mathbf{N}^{-1}\mathbf{M}^{-1}\mathbf{\Omega}^{-1}\begin{pmatrix}1\\0\\0\end{pmatrix} = \begin{pmatrix}\cos\omega\cos\mu\\ -\sin\omega\cos\nu+\cos\omega\sin\mu\sin\nu\\ \sin\omega\sin\nu+\cos\omega\sin\mu\cos\nu\end{pmatrix}. \quad (12)$$

With this definition, we can calculate $\mathbf{Q}_\nu$ for a set of observations. The monochromator and analyzer scattering angles provide E_i and E_f (and consequently k_i and k_f), which together with φ define q and θ according to equations (9) and (10). If we define the actual rotation angle about the instrument z axis to be s, we can write

$$s = \theta + \omega. \quad (13)$$

This allows for the calculation of ω, which together with μ and ν provides all the information needed to calculate $\mathbf{Q}_\nu$. The ability to calculate $\mathbf{Q}_\nu$ immediately allows us to determine the reciprocal-space coordinates from the spectrometer angles using

$$\mathbf{Q} = (\mathbf{UB})^{-1}\mathbf{Q}_\nu. \quad (14)$$

In addition, the calculation of this vector allows us to use the procedures of Busing & Levy (1967) for calculating the **U** matrix using either two non-collinear reflections with known lattice constants or three or more non-coplanar reflections with full refinement of the lattice constants.

2.1. Angle calculations for the triple-axis spectrometer

The problem of calculating angles for a given $\mathbf{Q} = (h, k, l)$, k_i and k_f is made more complicated by the fact that the problem is overdefined, as in the case of the four-circle diffractometer. This overdefinition arises from being able to use the sample manipulation angles to rotate the sample about **Q** producing many equivalent solutions. As a result, we need to impose some constraint to be able to calculate angles. However, the different sample goniometer used on the TAS means that the constraints discussed for the four-circle diffractometer are not particularly useful in the triple-axis case. As most triple-axis experiments are performed within, or at least in close proximity to, a fixed scattering plane, there are two particularly useful constraints that can be used:

(i) rotate **Q** into the scattering position so as to keep a specified reference plane as horizontal as possible;

(ii) minimize the deviation of the arcs from their zero position.

Mathematically, the first constraint corresponds to minimizing the angle between the specified reference plane normal vector and the instrument z axis. The second constraint corresponds to keeping the plane defined by $\mu = \nu = 0$ as 'level' as possible and, consequently, represents a specific example of case (i) above.

2.1.1. Keep a reference plane as horizontal as possible. We will begin this calculation with the instrument-angle matrix (Busing & Levy, 1967),

$$\mathbf{R} = \mathbf{\Omega}\mathbf{M}\mathbf{N}, \quad (15)$$

which transforms a vector from the ν coordinate system to the ω coordinate system. For the vector to be in the scattering position, we must have

$$\mathbf{R}\mathbf{Q}_\nu = \mathbf{Q}_\theta = \begin{pmatrix}q\\0\\0\end{pmatrix}. \quad (16)$$

To see how angles can be extracted from a specified value of the **R** matrix, we must first multiply the **Ω**, **M** and **N** matrices together, resulting in

$$\mathbf{R} = \begin{pmatrix}R_{11}&R_{12}&R_{13}\\R_{21}&R_{22}&R_{23}\\R_{31}&R_{32}&R_{33}\end{pmatrix} = \begin{pmatrix}\omega_\mathrm{c}\mu_\mathrm{c} & \omega_\mathrm{c}\mu_\mathrm{s}\nu_\mathrm{s}-\omega_\mathrm{s}\nu_\mathrm{c} & \omega_\mathrm{c}\mu_\mathrm{s}\nu_\mathrm{c}+\omega_\mathrm{s}\nu_\mathrm{s}\\ \omega_\mathrm{s}\mu_\mathrm{c} & \omega_\mathrm{s}\mu_\mathrm{s}\nu_\mathrm{s}+\omega_\mathrm{c}\nu_\mathrm{c} & \omega_\mathrm{s}\mu_\mathrm{s}\nu_\mathrm{c}-\omega_\mathrm{c}\nu_\mathrm{s}\\ -\mu_\mathrm{s} & \mu_\mathrm{c}\nu_\mathrm{s} & \mu_\mathrm{c}\nu_\mathrm{c}\end{pmatrix}, \quad (17)$$

where $\omega_\mathrm{c} = \cos\omega$, $\omega_\mathrm{s} = \sin\omega$, $\mu_\mathrm{c} = \cos\mu$, $\mu_\mathrm{s} = \sin\mu$, $\nu_\mathrm{c} = \cos\nu$ and $\nu_\mathrm{s} = \sin\nu$.

Given a value for **R**, we can extract values for μ, ν and ω from the following expressions:

$$\mu_\mathrm{s} = \sin\mu = -R_{31},\qquad \mu_\mathrm{c} = \cos\mu = (R_{11}^2 + R_{21}^2)^{1/2}, \quad (18)$$

$$\nu_\mathrm{s} = \sin\nu = R_{32}/(R_{11}^2 + R_{21}^2)^{1/2},\qquad \nu_\mathrm{c} = \cos\nu = R_{33}/(R_{11}^2 + R_{21}^2)^{1/2}, \quad (19)$$

$$\omega_\mathrm{s} = \sin\omega = R_{21}/(R_{11}^2 + R_{21}^2)^{1/2},\qquad \omega_\mathrm{c} = \cos\omega = R_{11}/(R_{11}^2 + R_{21}^2)^{1/2}. \quad (20)$$

The only quadrant restriction that results from this calculation is that the upper arc, μ, is restricted to lie between $\pm 90^\circ$ because of the expression for $\cos\mu$. However, the configuration of the typical goniometer for a triple-axis spectrometer restricts the motion of both arcs to be less than $\pm 25^\circ$ and, consequently, this restriction is not relevant.

If we can now calculate the value of **R** so as to minimize the angle between the reference plane normal and the z axis of the instrument, we have completed our goal. In practice, the reference plane can be defined by specifying a plane normal, specifying a pair of vectors within the plane or specifying a set of arc values. All three manners of specifying the plane can be equivalently expressed by the plane normal vector in the ν coordinate system. To see how the plane normal vector is related to the pair of arc values that define the plane (which we will refer to as μ_plane and ν_plane) we can write out the expression for the plane normal in the ν coordinate system by first considering a vector along the z axis in the laboratory coordinate system and multiplying by the appropriate rotation matrices,

$$\mathbf{u}_{\nu\perp} = \mathbf{N^{-1}M^{-1}}\begin{pmatrix}0\\0\\1\end{pmatrix} = \begin{pmatrix}-\sin\mu_{\rm plane}\\ \cos\mu_{\rm plane}\sin\nu_{\rm plane}\\ \cos\mu_{\rm plane}\cos\nu_{\rm plane}\end{pmatrix}. \quad (21)$$

We will now transform the **Q** of interest into the ν coordinate system using the known **U** and **B** matrices,

$$\mathbf{Q}_\nu = \mathbf{UBQ}. \quad (22)$$

Normalizing this vector gives us a unit vector along the direction of $\mathbf{Q}_\nu$, which we define to be $\mathbf{u}_{1\nu}$. If we take the cross-product of $\mathbf{u}_{\nu\perp}$ and $\mathbf{u}_{1\nu}$, we can define a new vector $\mathbf{u}_{2\nu}$,

$$\mathbf{u}_{2\nu} = \mathbf{u}_{\nu\perp} \times \mathbf{u}_{1\nu}. \quad (23)$$

This vector has the essential properties that it is normal to $\mathbf{u}_{\nu\perp}$, which places it in the reference plane, and it is also normal to **Q**. Consequently, if we can rotate **Q** into the scattering position about the direction defined by $\mathbf{u}_{2\nu}$, the angular deviation between the instrument z axis and the reference plane normal will be minimized.

To perform this rotation, we will define a new orthonormal set using these three vectors as

$$\begin{aligned}\widehat{\mathbf{t}}_{1\nu} &= \mathbf{u}_{1\nu},\\ \widehat{\mathbf{t}}_{2\nu} &= \mathbf{u}_{2\nu},\\ \widehat{\mathbf{t}}_{3\nu} &= \widehat{\mathbf{t}}_{1\nu} \times \widehat{\mathbf{t}}_{2\nu}.\end{aligned} \quad (24)$$

Note that we form the vector $\widehat{\mathbf{t}}_{3\nu}$ as vectors $\mathbf{u}_{1\nu}$ and $\mathbf{u}_{\nu\perp}$ are not necessarily perpendicular to one another. The matrix **R** must now transform the vectors $\widehat{\mathbf{t}}_{1\nu}$, $\widehat{\mathbf{t}}_{2\nu}$ and $\widehat{\mathbf{t}}_{3\nu}$ from the ν coordinate system into the θ coordinate system, such that

$$\mathbf{R}\widehat{\mathbf{t}}_{1\nu} = \begin{pmatrix}1\\0\\0\end{pmatrix}, \quad (25)$$

$$\mathbf{R}\widehat{\mathbf{t}}_{2\nu} = \begin{pmatrix}0\\1\\0\end{pmatrix}, \quad (26)$$

$$\mathbf{R}\widehat{\mathbf{t}}_{3\nu} = \begin{pmatrix}0\\0\\1\end{pmatrix}. \quad (27)$$

Equation (25) ensures that **Q** is in the scattering condition, while equation (26) ensures that a vector normal to **Q** yet contained within the reference plane is also in the horizontal scattering plane. Note that we can rewrite this expression in matrix form by forming the matrix $\mathbf{T}_\nu$, which has vectors $\widehat{\mathbf{t}}_{1\nu}$, $\widehat{\mathbf{t}}_{2\nu}$, and $\widehat{\mathbf{t}}_{3\nu}$ as its columns. We can rewrite the above three equations as

$$\mathbf{RT}_\nu = \begin{pmatrix}1&0&0\\0&1&0\\0&0&1\end{pmatrix}, \quad (28)$$

and therefore

$$\mathbf{R} = \mathbf{T}_\nu^{-1}. \quad (29)$$

Note that the only information that went into this calculation was the reciprocal-lattice coordinate, which we wish to place in the scattering position, and the reference plane arcs. The key point in this calculation is that at the end of the transformation, **Q** is in the scattering position and a vector normal to **Q** contained within the reference plane is also in the horizontal scattering plane. Therefore, by definition **Q** was rotated into the scattering plane so as to keep the reference plane as 'level' as possible, which was precisely our original goal.

With the **R** matrix calculated, we can use equations (18), (19) and (20) to calculate values for ω, μ and ν. The full set of angles we wish to calculate would be the set that would place a specified $\mathbf{Q} = (h, k, l)$ into the scattering position with a specified $E_{\rm i}$ and $E_{\rm f}$. The values for $E_{\rm i}$ and $E_{\rm f}$ can be used to calculate $k_{\rm i}$ and $k_{\rm f}$ and, consequently, the monochromator and analyzer scattering angles. The magnitude of **Q** together with $k_{\rm i}$ and $k_{\rm f}$ can be used to calculate φ according to (9). These values can then be used to calculate θ from (10), which, together with the value for ω resulting from the calculation described above, can be used to calculate the value for the observed rotation angle s using (13). These angles together with the arc values from the calculation above completely define the required set of angles.

As mentioned above, the second mode of calculation is minimizing the absolute value of the arcs. This mode is simply a special case of the mode described above with the reference plane arcs set to zero, *i.e.* $\mu_{\rm plane} = \nu_{\rm plane} = 0$.

This completes the extension of the UB matrix formalism to handle inelastic scattering experiments using the typical geometry of the triple-axis spectrometer. As a final note, this formalism has been implemented in our new *LabVIEW*-based data acquisition system, *SPICE* (http://neutron.ornl.gov/spice) used for controlling the neutron scattering instruments at the High-Flux Isotope Reactor. This program has been used extensively by both local staff and external users and has greatly enhanced the capabilities of the triple-axis spectrometers.

3. Time-of-flight spectrometers

Inelastic neutron scattering measurements are also made using time-of-flight (TOF) spectroscopy. Such measurements use a pulsed beam of neutrons and the measured time taken for the neutron to travel from the sample to the detector determines the energy transfer. These spectrometers are of two types, corresponding to fixed $E_{\rm i}$ with varying $E_{\rm f}$ determined by time-of-flight (referred to as direct geometry) or fixed $E_{\rm f}$ with varying $E_{\rm i}$ (referred to as indirect geometry). In either case, an individual measurement corresponds to some $\mathbf{k}_{\rm i}$ and $\mathbf{k}_{\rm f}$ scattered into a detector pixel. The primary difference between this and the case of the triple-axis spectrometer is that the detector location, and thus the direction of $\mathbf{k}_{\rm f}$, is in a general spatial position, *i.e.* it is not restricted to be in a plane.

As in the case of the triple-axis spectrometer, we consider a neutron beam incident along the y axis. The direction of $\mathbf{k}_{\rm f}$ is defined to be at an angle φ' from the y axis when projected on to the xy plane and angle ψ out of this plane. From this definition of angles, shown in Fig. 3, we can write **Q** in the laboratory coordinate system as

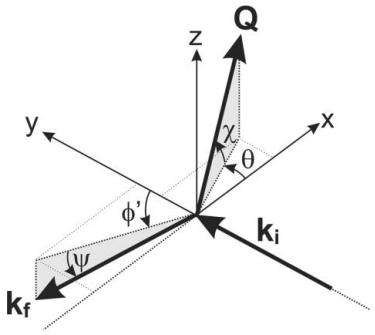

Figure 3
Coordinate system for TOF spectrometers. The direction of $\mathbf{k}_f$ is at an angle φ' from the y axis when projected onto the xy plane and ψ away from this plane ($\mathbf{k}_f$ is shown rotated below the xy plane in the above figure). This results in $\mathbf{Q}$ at an angle χ out of the xy plane with an xy plane projection rotated by θ from the x axis.

$$\begin{aligned}\mathbf{Q}_L &= \mathbf{k}_i - \mathbf{k}_f \\ &= \begin{pmatrix} 0 \\ k_i \\ 0 \end{pmatrix} - \begin{pmatrix} -k_f \cos\psi \sin\varphi' \\ k_f \cos\psi \cos\varphi' \\ -k_f \sin\psi \end{pmatrix} \\ &= \begin{pmatrix} k_f \cos\psi \sin\varphi' \\ k_i - k_f \cos\psi \cos\varphi' \\ k_f \sin\psi \end{pmatrix}. \end{aligned} \tag{30}$$

If we split $\mathbf{Q}_L$ into components within and perpendicular to the xy plane,

$$Q_L^2 \equiv q^2 = Q_\parallel^2 + Q_\perp^2, \tag{31}$$

we can use (30) and the angle definitions shown in Fig. 3 to write

$$Q_\parallel = (k_i^2 - 2k_i k_f \cos\psi \cos\varphi' + k_f^2 \cos^2\psi)^{1/2}, \tag{32}$$

$$Q_\perp = k_f \sin\psi,$$

$$\tan\theta = (k_i - k_f \cos\psi \cos\varphi')/k_f \cos\psi \sin\varphi',$$

$$\tan\chi = (k_f \sin\psi)/Q_\parallel.$$

Note that the we can easily relate the true scattering angle (which we will refer to as φ in direct analogy with the triple-axis case) to the angles defined in Fig. 3. To see this relation, we will first write

$$\begin{aligned} Q_L \equiv q &= (Q_\parallel^2 + Q_\perp^2)^{1/2} \\ &= (k_i^2 + k_f^2 - 2k_i k_f \cos\psi \cos\varphi')^{1/2}. \end{aligned} \tag{33}$$

In addition, the law of cosines allows us to write an expression for $\mathbf{Q}_L$ using the true scattering angle, φ, as

$$Q_L \equiv q = (k_i^2 + k_f^2 - 2k_i k_f \cos\varphi)^{1/2}, \tag{34}$$

which is identical to the expression we had in the triple-axis case [equation (9)]. Equating (33) with (34) allows us to write a simple relation between φ and the angles defined in Fig. 3, φ' and ψ,

$$\cos\varphi = \cos\psi \cos\varphi'. \tag{35}$$

There is no standard goniometer used in TOF spectrometers and so, for direct comparison purposes, we will assume the same set of angles used in the triple-axis case. This assumption allows us to write the same scattering equation for this case:

$$\mathbf{Q}_L = \mathbf{\Theta\Omega MNUBQ}. \tag{36}$$

As before, we can write out the equation in the θ coordinate system by multiplying $\mathbf{Q}_L$ by $\mathbf{\Theta}^{-1}$, which gives

$$\mathbf{Q}_\theta = \begin{pmatrix} (k_i^2 - 2k_i k_f \cos\psi \cos\varphi' + k_f^2 cos^2\psi)^{1/2} \\ 0 \\ k_f \sin\psi \end{pmatrix}, \tag{37}$$

$$\mathbf{Q}_\theta = \begin{pmatrix} Q_\parallel \\ 0 \\ Q_\perp \end{pmatrix}. \tag{38}$$

We can compare this expression with the expression for $\mathbf{Q}_\theta$ for the triple-axis spectrometer [equation (11)]. In the case of the TOF spectrometer, the general spatial position of the detector results in $\mathbf{Q}_\theta$ lying in the xz plane with the in-plane component along the x axis and the out-of-plane component along the z axis.

We can now write the expression for $\mathbf{Q}$ in the ν coordinate system by multiplying by the appropriate rotation matrices,

$$\begin{aligned}\mathbf{Q}_\nu &= \mathbf{N}^{-1}\mathbf{M}^{-1}\mathbf{\Omega}^{-1}\mathbf{Q}_\theta \\ &= \begin{pmatrix} Q_\parallel \omega_c \mu_c - Q_\perp \mu_s \\ -Q_\parallel \omega_s \nu_c + Q_\parallel \omega_c \mu_s \nu_s + Q_\perp \mu_c \nu_s \\ Q_\parallel \omega_s \nu_s + Q_\parallel \omega_c \mu_s \nu_c + Q_\perp \mu_c \nu_c \end{pmatrix}, \end{aligned} \tag{39}$$

where $\omega_c = \cos\omega$, $\omega_s = \sin\omega$, $\mu_c = \cos\mu$, $\mu_s = \sin\mu$, $\nu_c = \cos\nu$ and $\nu_s = \sin\nu$.

If we normalize by the magnitude of $\mathbf{Q}$, we can define

$$\widehat{Q}_\parallel = Q_\parallel/(Q_\parallel^2 + Q_\perp^2)^{1/2}, \tag{40}$$

$$\widehat{Q}_\perp = Q_\perp/(Q_\parallel^2 + Q_\perp^2)^{1/2},$$

which allows us to write an expression for a unit vector in the ν coordinate system as

$$\mathbf{u}_\nu = \begin{pmatrix} \widehat{Q}_\parallel \omega_c \mu_c - \widehat{Q}_\perp \mu_s \\ -\widehat{Q}_\parallel \omega_s \nu_c + \widehat{Q}_\parallel \omega_c \mu_s \nu_s + \widehat{Q}_\perp \mu_c \nu_s \\ \widehat{Q}_\parallel \omega_s \nu_s + \widehat{Q}_\parallel \omega_c \mu_s \nu_c + \widehat{Q}_\perp \mu_c \nu_c \end{pmatrix}. \tag{41}$$

As a consistency check, we can consider the planar case. Restricting the detectors to lie in the xy plane results in $\psi \to 0$, $\widehat{Q}_\perp \to 0$ and $\widehat{Q}_\parallel \to 1$. This limit results in the same expression as we had for the TAS case [equation (12)].

A single measurement consists of a scattering event with a specified k_i and k_f together with the angular coordinates specifying the direction of $\mathbf{k}_f$ and the sample orientation angles. The values of k_i, k_f, φ' and ψ allow us to calculate $Q_\parallel$, $Q_\perp$ and θ according to equation (32). The calculation of θ together with the observed rotation angle allows us to calculate ω [equation (13)], which together with μ and ν provides all the information needed to calculate Q_ν using (39). There-

fore, we can directly convert an observed scattering event into reciprocal-space coordinates using (14). In addition, we can use the procedures outlined by Busing & Levy (1967) to calculate the **U** matrix.

3.1. Angle calculations for TOF spectrometers

The general case of calculating a set of angles for the case of the TOF spectrometer is difficult because we have an added degree of freedom over the case of the triple-axis as the detectors are not planar. Therefore, we will consider two calculations that should be very useful given the way that TOF spectrometers are currently used:

(i) calculating the arc angles required to place a specified plane horizontal;

(ii) calculating angles for the case where the only sample manipulation is achieved through a rotation stage (*i.e.* $\mu = \nu = 0$).

3.1.1. Angles required to place a plane horizontal. Frequently, inelastic neutron scattering experiments are performed with the sample aligned with a specific plane horizontal. Even with the large detector coverage of many TOF spectrometers, there are advantages to having a sample accurately aligned in a highly symmetric geometry. Consequently, the presence of a set of orientation arcs, such as those typically employed on a triple-axis spectrometer, would quite often be used to adjust the sample alignment to align a plane accurately. As a result, we will show the calculations necessary to place a plane horizontal.

We will begin with some specified scattering plane. This can be specified using either a pair of vectors within the plane or the plane normal. Clearly, the former can easily be converted into the latter and we will assume that the plane is defined by a plane normal unit vector. This vector, represented in the ν coordinate system, $\mathbf{u}_{\nu\perp}$, will be defined as

$$\mathbf{u}_{\nu\perp} = \begin{pmatrix} u_{1\nu\perp} \\ u_{2\nu\perp} \\ u_{3\nu\perp} \end{pmatrix}. \tag{42}$$

The goal of our calculation is to have $\mathbf{u}_{\nu\perp}$ vertical in the laboratory coordinate system, which means we want

$$\mathbf{Q}_{\rm L}/|\mathbf{Q}_{\rm L}| = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} = \mathbf{\Theta\Omega MN u}_{\nu}. \tag{43}$$

From this, we can write the expression for $\mathbf{u}_{\nu}$ that will place a vector vertical in the laboratory coordinate system:

$$\mathbf{u}_{\nu} = \mathbf{N^{-1}M^{-1}}\begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} = \begin{pmatrix} -\sin\mu \\ \cos\mu\sin\nu \\ \cos\mu\cos\nu \end{pmatrix}. \tag{44}$$

If we equate equations (44) and (42), we can write the following expressions for μ and ν:

$$\begin{aligned} u_{1\nu\perp} &= -\sin\mu, \\ u_{2\nu\perp}/u_{3\nu\perp} &= \tan\nu. \end{aligned} \tag{45}$$

As both μ and ν are restricted to lie between $\pm 90^\circ$, the above expressions uniquely describe the set of arcs required to place the specified plane horizontal.

3.1.2. Case of no goniometer arcs. The majority of TOF spectrometers in use today have only the ability to manipulate the sample orientation through a single rotation stage with a vertical axis of rotation. This corresponds to the case where $\mu = \nu = 0$. In such a restricted geometry, the ability to rotate about the scattering vector is eliminated and the problem of calculating angles is no longer overdefined. To see how angles are calculated in this case, we start with the expression for **Q** in the ν coordinate system [equation (39)]. Setting the values of μ and ν to zero in this expression gives

$$\mathbf{Q}_{\nu} = \begin{pmatrix} Q_{\parallel}\cos\omega \\ -Q_{\parallel}\sin\omega \\ Q_{\perp} \end{pmatrix}. \tag{46}$$

Therefore, if we want to drive a specific $\mathbf{Q} = (h, k, l)$ into the scattering position for a specified $k_{\rm i}$ and $k_{\rm f}$, we must first transform **Q** into the ν coordinate system,

$$\mathbf{Q}_{\nu} = \mathbf{UBQ} \equiv \begin{pmatrix} Q_{1\nu} \\ Q_{2\nu} \\ Q_{3\nu} \end{pmatrix}. \tag{47}$$

Equating equations (46) and (47), immediately allows us to write

$$Q_{\perp} = Q_{3\nu} = k_{\rm f}\sin\psi, \tag{48}$$

which defines the out-of-plane location of $\mathbf{k}_{\rm f}$ as defined by the angle ψ. As the ν coordinate system is a valid orthonormal coordinate system, we can determine $|\mathbf{Q}|$ from

$$|\mathbf{Q}|^2 = q^2 = Q_{1\nu}^2 + Q_{2\nu}^2 + Q_{3\nu}^2, \tag{49}$$

which together with equations (31) and (48) gives $Q_{\parallel}$. We can use $Q_{\parallel}$ to define ω uniquely from

$$\begin{aligned} \cos\omega &= Q_{1\nu}/Q_{\parallel}, \\ \sin\omega &= -Q_{2\nu}/Q_{\parallel}. \end{aligned} \tag{50}$$

We can now use $k_{\rm i}$, $k_{\rm f}$, ψ and the expression for $Q_{\parallel}$ to write

$$\cos\varphi' = \frac{k_{\rm i}^2 + k_{\rm f}^2\cos^2\psi - Q_{\parallel}^2}{2k_{\rm i}k_{\rm f}\cos\psi}. \tag{51}$$

Under the assumption that ψ is always between $\pm 90^\circ$ ($\cos\psi > 0$) we can solve for φ' as above. Note that we have a choice of sign for φ' in the above expression. The choice of the sign for φ' will determine the value for θ according to (32). The actual orientation angle, s, is now determined from (13) and thus the full set of required angles can be obtained.

4. Summary

In summary, we have extended the UB matrix approach of Busing & Levy (1967) to handle the case of inelastic neutron scattering. We have considered the standard inelastic neutron scattering instruments for both continuous and pulsed neutron sources, namely the triple-axis spectrometer and the TOF spectrometer. For the conventional sample goniometer, we

have developed expressions that allow for the calculation of the UB matrix and the conversion from angles to h, k, l, E space. In addition, we have developed schemes for the calculation of angles for a given h, k, l, k_i and k_f. We have been using this procedure for some time now on the triple-axis spectrometers at the High-Flux Isotope Reactor and this approach has greatly improved the capability of this rather mature instrument. Although not currently being used on TOF spectrometers, such an approach should drastically improve the single-crystal handling capabilities of such instruments and should have a huge impact on the quality of science performed on such an instrument.

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